The 0 and the π phase Josephson coupling through an insulating barrier with magnetic impurities

O. Vávra,[1, 2, *] S. Gaži,[2] D. S. Golubović,[1] I. Vávra,[2] J. Dérer,[2]
J. Verbeeck,[3] G. Van Tendeloo,[3] and V. V. Moshchalkov[1]

[1]INPAC-Institute for Nanoscale Physics and Chemistry,
Nanoscale Superconductivity and Magnetism Group,
K. U. Leuven, Celestijnenlaan 200 D, B-3001 Leuven, Belgium
[2]Institute of Electrical Engineering, Slovak Academy of Sciences,
Dúbravská cesta 9, SK-841 04 Bratislava, Slovak Republic
[3]EMAT, University of Antwerp, Groenenborgerlaan 171, B-2020, Antwerp, Belgium
(Dated: February 6, 2008)

We have studied temperature and field dependencies of the critical current I_C in the Nb-Fe$_{0.1}$Si$_{0.9}$-Nb Josephson junction with tunneling barrier formed by paramagnetic insulator. We demonstrate that in these junctions the co-existence of both the 0 and the π states within one tunnel junction takes place which leads to the appearance of a sharp cusp in the temperature dependence $I_C(T)$ similar to the $I_C(T)$ cusp found for the $0 - \pi$ transition in metallic π junctions. This cusp is not related to the $0 - \pi$ temperature induced transition itself, but is caused by the different temperature dependencies of the opposing 0 and π supercurrents through the barrier.

PACS numbers: 74.50.+r, 85.25.Cp

As first predicted by Josephson[1], the supercurrent I_S through the tunnel barrier is driven by the phase difference φ across the junction applied to the superconducting wave function. In conventional Josephson junctions (JJ) this current is described by the relation $I_S = I_C sin\varphi$, where I_C is the critical current. Recently, a considerable attention has been devoted to the investigation of the π JJs[2,3,4]. In this case the relation between the supercurrent and the phase difference is $I_S = I_C sin(\varphi + \pi) = -I_C sin\varphi$[5]. One of the possible realizations of the π junctions is the superconductor-ferromagnetic metal-superconductor (S-FM-S) tunnel junction, wherein the spatial oscillations of the superconducting order parameter occur in the ferromagnetic metal as a consequence of the exchange splitting of the conduction band[6]. The transition between the 0 and the π states was experimentally observed as the vanishing of the Josephson current. The $0 - \pi$ transition can be induced by the varying barrier thickness[2,4] or the temperature[3,4]. As the absolute value of the current is measured, e.g. for proper values of the ferromagnetic barrier thickness, a sharp cusp in the temperature dependence of the critical current $I_C(T)$ is observed as a consequence of the $0 - \pi$ transition.

It is also predicted that JJs with magnetic impurities within an *insulating barrier* can produce the π state[5]. Later on, the possibilities to observe the π junctions in JJs with ferromagnetic insulating or semiconducting barrier (S-FI-S) were analyzed theoretically in[7,8]. In such types of JJs the proximity effect in the barrier is much weaker, as compared with the ferromagnetic metal, and can be disregarded. In this case the formation of the π junction is caused by the quasiparticle scattering on a magnetically active interfaces[8]. It can result in the splitting of the Andreev interface bound-state energies into two spin channels[7]. Theoretically, if these channels com-

pensate each other the $0 - \pi$ transition is observed. Up to now the π state in the JJs with insulating magnetic barrier has not been found experimentally.

In this letter we present the experimental evidence of the existence of the π state in the Josephson junction with magnetic impurities in the insulating barrier. We also demonstrate that the co-existence of both the 0 and the π states within one tunnel junction leads to the appearance of a similar cusp in the temperature dependence of I_C as the one for the $0-\pi$ transition in metallic π junctions. The origin of this cusp, however, is not related to the $0 - \pi$ transition itself, but rather to a simultaneous 0 and π Josephson tunneling through an insulating barrier
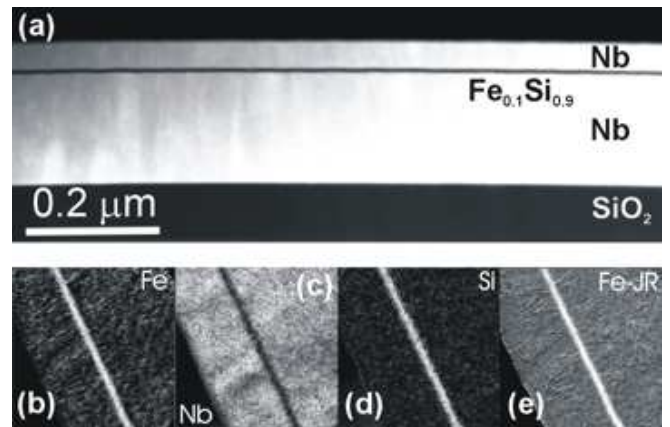

FIG. 1: Cross-sectional micrographs of the Nb-Fe$_{0.1}$Si$_{0.9}$-Nb tunnel junction obtained in various modes of transmission electron microscope: (a) z-contrast annular dark-field micrograph, (b),(c) and (d) energy filtered TEM elemental maps of Fe, Nb and Si, (e) jump-ratio Fe elemental map. The thickness of the Fe$_{0.1}$Si$_{0.9}$ barrier is 6 nm.
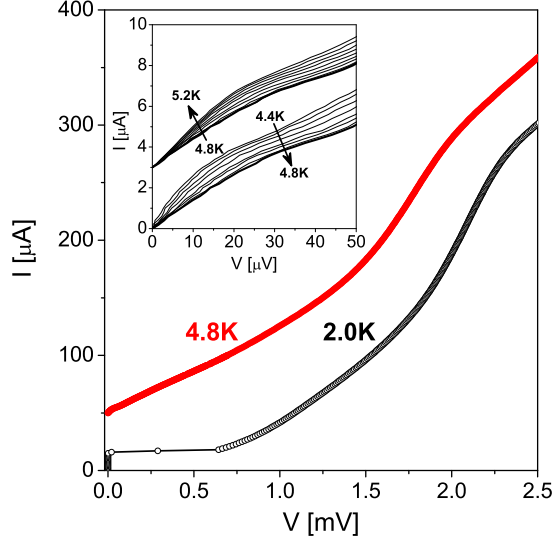
FIG. 2: Current-voltage characteristics of the Nb-Fe$_{0.1}$Si$_{0.9}$-Nb tunnel junction at 4.8 K and 2.0 K. The curves are shifted by $50\mu A$ for clarity. Inset: The temperature dependent change of the slope of the IV curves around 4.8 K. The curves for the range from 4.8 K to 5.2 K are shifted by $3\mu V$ for clarity.

with magnetic impurities. In this case, quite generally, the tunneling through an insulating barrier itself gives rise to a positive contribution into the critical current I_{C0} (0-part), whereas the tunneling via scattering on magnetic impurities generates a negative $I_{C\pi}$ (π-part). These two currents have opposite signs and different temperature dependencies of the critical currents, which results in their complete mutual cancelation $|I_{C0} - I_{C\pi}| = 0$ at a certain temperature where a sharp depression of the critical current has been observed.

We have studied a Nb-Fe$_{0.1}$Si$_{0.9}$-Nb tunnel junction, with the amorphous Fe$_{0.1}$Si$_{0.9}$ alloy as the barrier. Amorphous magnetic materials here have certain advantages compared to polycrystalline materials, because of a lack of crystalline defects and a better composition homogeneity at a microscopic level. Moreover, Fe$_{0.1}$Si$_{0.9}$ alloy is additionally favorable, since it is an insulator at low temperatures, with the resistance a few orders of magnitude higher than that of the metallic alloys. The Fe$_{0.1}$Si$_{0.9}$ alloy is a paramagnetic material[9], but the amorphous structure does not rule out completely the possible existence of a local ferromagnetic exchange field at low temperatures. The molecular dynamics ab-initio simulation reveals that nearest neighbor (NN) positions of the Fe atoms are also quite probable. From that point of view, the formation of microscopic regions with ferromagnetic exchange coupling seems to be possible. In this case the barrier can be thought of as a "nanocomposite", containing regions with and without magnetic exchange coupling.
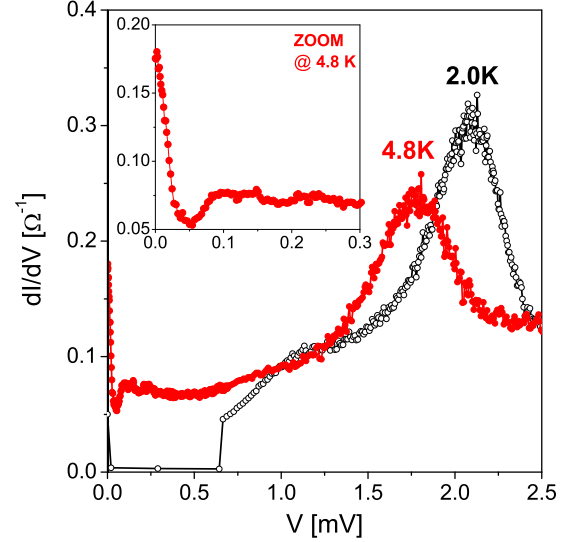

FIG. 3: Differential conductance vs. bias voltage characteristics of the Nb-Fe$_{0.1}$Si$_{0.9}$-Nb junctions at 4.8 K and 2.0 K. Inset: Zoom, showing the zero-bias conductance peak at 4.8 K.

The Nb-Fe$_{0.1}$Si$_{0.9}$-Nb junctions were prepared by a sputtering technique under the conditions similar to those used for the fabrication of the Nb-Si-Nb junctions[10]. The area of the junction is $20 \times 20\mu m^2$. The structure and composition homogeneity of the 6 nm thick barrier were investigated by transmission electron microscopy (TEM) in a cross-sectional specimen (fig. 1). These studies have clearly confirmed that the barrier is very well defined. Indications of neither strong interdiffusion nor local Nb shorts were found. The Fe$_{0.1}$Si$_{0.9}$ barrier is very homogeneous in thickness as well as in composition.

The current-voltage characteristics (IV) of the Nb-Fe$_{0.1}$Si$_{0.9}$-Nb junction were measured (Fig. 2), and subsequently the differential conductance (dI/dV) versus the bias voltage was determined numerically (Fig. 3). At both temperatures 4.8 K and 2 K peaks are observed at voltages $V = 1.77mV$ and $V = 2.09mV$, respectively. They correspond to the sum of the superconducting gaps related to the individual Nb electrodes of the tunnel junction. A reduced value of the sum of the superconducting gaps is most probably due to a non-ideal upper Nb electrode (see below).

At the temperature of 2K, the critical current is $I_C = 17\mu A$. As the I_C value is finite, the derivative of the current-voltage curve at the zero bias is infinite. Such IV and dI/dV curves are typical for the temperatures and magnetic fields where the junction has a finite I_C value.

The dI/dV curve measured at 4.8 K corresponds to the applied magnetic flux $\Phi/\Phi_0 = 0.7$ where the maximum of the zero-bias peak was observed [see Fig. 5]. Such type of the dI/dV curves is typical for the temperatures and magnetic fields where the measurable critical current is
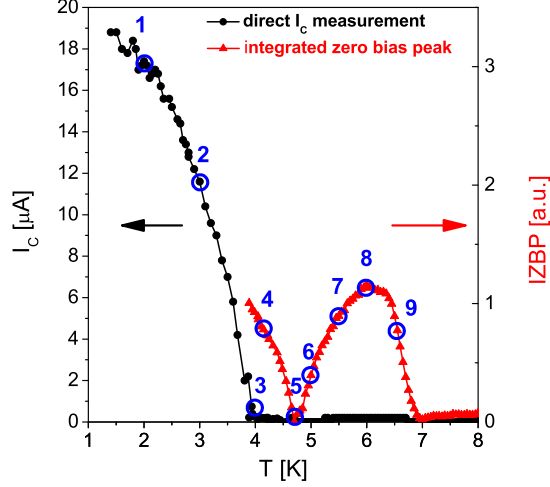
FIG. 4: The temperature dependencies of the critical current $I_C(T)$ and integrated zero-bias peak $IZBP(T)$ of the Nb-Fe$_{0.1}$Si$_{0.9}$-Nb tunnel junction in the zero magnetic field. The $IZBP(T)$ can be considered as a zoom of the $I_C(T)$ dependence in the temperature range from 4 K up to 7 K. The circles marks denotes the points where the $I_C(\Phi)$ was measured [see Fig. 5].

absent.

For the reference junction Nb-Si-Nb[10] the upper Nb electrode contains a thin ($\sim$ 2 nm) sublayer of amorphous Nb adjacent to the silicon barrier. This junction behaves like the SINS system, where N represents the amorphous part of the Nb electrode. Due to the non-equal atomic condensation of Nb on Fe$_{0.1}$Si$_{0.9}$, similar amorphous Nb sublayer was also found in the Nb-Fe$_{0.1}$Si$_{0.9}$-Nb junctions. The absence of the measurable critical current at zero bias is caused by the decay of the superconducting order parameter in this amorphous part of the Nb electrode as well as by the Andreev scattering at the interface of the polycrystalline and amorphous Nb. Similar zero-bias peak in dI/dV was described by Klapwijk[11] and in his case of the Nb-Si-Nb junction was considered as a precursor of the fully developed supercurrent observed for thinner barriers. In our case the zero-bias peak in dI/dV can be interpreted as a precursor of the supercurrent for thinner amorphous part of the upper Nb electrode. To obtain the information about the precursor of the I_C we introduced the integrated zero-bias peak ($IZBP$) amplitude

$$IZBP = \int_0^{V_C} (dI/dV(V) - dI/dV_{offset})dV, \quad (1)$$

where V_C is a voltage criterion (we used $V_C = 5\mu V$), and dI/dV_{offset} is the reference conductance value. From the $IZBP(T)$ and $IZBP(\Phi)$ data (see below) we confirm that the zero-bias peak is the precursor of I_C.
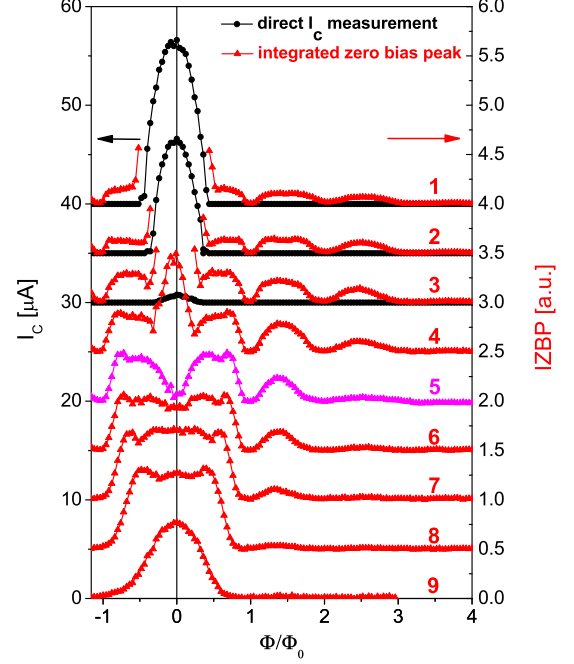

FIG. 5: The applied magnetic flux dependencies of the critical current $I_C(\Phi)$ and integrated zero-bias peak $IZBP(\Phi)$ of the Nb-Fe$_{0.1}$Si$_{0.9}$-Nb tunnel junction for temperatures marked in the Fig. 4.

Direct measurements of the I_C at the zero magnetic field reveal some finite values up to the temperature around 4 K [see Fig. 4]. Above this temperature, instead of I_C the zero bias-peak is observed in the dI/dV curves. To find out what happens above 4 K, we have determined the $IZBP$ for each used temperature $T > 4K$. As it can be seen from Fig. 4, the proposed method reveals a sharp $IZBP$ cusp at the temperature of 4.8 K [for correlation see also the temperature dependent change of the slope of the IV curves around the zero bias in the inset in Fig. 2], the maximum at approximately 6 K, and then decrease down to zero at $T \approx 7K$ which is the critical temperature of our Nb-Fe$_{0.1}$Si$_{0.9}$-Nb JJ.

Fig. 5 shows the critical current vs. applied magnetic flux $I_C(\Phi)$ dependencies for the temperatures marked in Fig. 4 by circles. Curves 1, 2 and 3 show the peak around the zero field with finite values of I_C. Elsewhere, the $I_C(\Phi)$ dependence is suppressed and was, therefore, obtained using $IZBP$ versus the applied magnetic flux. Similarly to $IZBP$ versus the temperature, the $IZBP$ was found for each value of the magnetic flux. In what follows the $IZBP$ together with the I_C temperature and magnetic field dependencies will be referred to as a single $I_C(T)$ or $I_C(\Phi)$ dependence.

The unusual behavior of the junction in magnetic field is clearly seen from Fig. 5. As the temperature decreases, the shape of the $I_C(\Phi)$ changes. Especially, in the interval of magnetic flux $\Phi \in \langle -\Phi_0; \Phi_0 \rangle$ it is visible that the
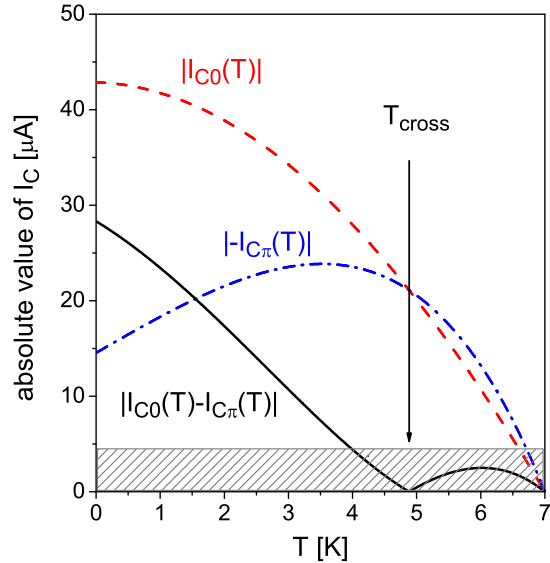
behaviour and a lack of electron diffraction rings typical for nanocrystallites in an amorphous matrix, the latter scenario seems to be less probable.

In our case the co-existence of the 0 and π junctions can be simulated as a JJ with a nonuniform spatial distribution of the critical current density and with additional polarity alternations. The assumption about the simultaneous presence of the 0 and the π tunneling is confirmed by the unusual shape of the $I_C(\Phi)$ curves (Fig. 5). When both 0 and π phases of the Josephson supercurrent co-exist in one Nb-Fe$_{0.1}$Si$_{0.9}$-Nb junction, two $I_C(T)$ dependencies must be taken into account: $I_{C0}(T)$ and $I_{C\pi}(T)$ (Fig. 6). Due to the lack of the adequate theories for such kind of the junctions the $I_{C0}(T)$ and the $I_{C\pi}(T)$ dependencies were calculated by using the theory of the π junctions with metallic barrier[4,15]. In this illustrative simulation the $I_{C0}(T)$ and the $I_{C\pi}(T)$ are taken for the same barrier thickness but with different values of the ferromagnetic exchange energy, the decay length ξ_{F1}, and oscillation period of the order parameter $2\pi\xi_{F2}$ (details will be provided elsewhere). The sum of these two currents of the opposite polarities (positive for I_{C0} and negative for $I_{C\pi}$) gives the $I_C(T)$ dependence which is similar to the one we have found (compare Fig. 4 and Fig. 6). The minimum of the $I_C(T)$ dependence T_{cross} corresponds to the crossing point of the $|-I_{C\pi}(T)|$ and $|I_{C0}(T)|$ dependencies [see Fig. 6].

In conclusion, we have observed the co-existence of the 0 and the π state in the Nb-Fe$_{0.1}$Si$_{0.9}$-Nb Josephson junctions with a paramagnetic insulating barrier formed by an amorphous Fe$_{0.1}$Si$_{0.9}$. Different temperature dependencies of the I_{C0} and $I_{C\pi}$ currents and their opposite signs lead to the appearance of very sharp cusp in the $I_C(T)$ curve at about 4.8 K where these two currents cancel each other completely. The simultaneous presence of both the 0 and the π phases in the Nb-Fe$_{0.1}$Si$_{0.9}$-Nb junction has been interpreted in terms of the "vortex state" model proposed by Bulaevskii for JJs with an insulating barrier with magnetic impurities. The adequate detailed theory which fully describes our experimental data is currently lacking and further interactions between theory and experiment are needed to reveal the nature of the phase-shifting effect in JJs with an insulating paramagnetic barrier.

FIG. 6: Simplified illustrative model of the Nb-Fe$_{0.1}$Si$_{0.9}$-Nb junction: The theoretical partial temperature dependencies of the critical current for the 0- and π-parts and their sum, showing the cusp in the $I_C(T)$ caused by crossing of the $I_{C0}(T)$ and $I_{C\pi}(T)$ curves. Shaded area: The levels of the I_C measured by IZBP due to the absence of the measurable critical current.

middle peak gradually vanishes as the temperature increases (curves 1-4). At the temperature of 4.8 K (curve 5) the minimum of the critical current at zero applied flux $I_C(0)$ is observed. Such behavior with the minimum of critical current at $\Phi = 0$ is typical for the $0 - \pi$ JJs[12]. Then for the temperatures $4.8K < T < 6K$ the shape of the $I_C(\Phi)$ curves is changing again and the $I_C(0)$ recovers its zero field maximum (curves 6-9). It is worth noting that in a reference Nb-Si-Nb JJ the $I_C(\Phi)$ shows a well defined conventional Fraunhofer like patterns[13]

As predicted Bulaevskii[12] a perfectly flat and homogeneous insulating barrier with magnetic impurities can induce the formation of an admixture of the π and 0 junctions ("vortex states") for certain range of the barrier parameters and temperatures. Since the $0 - \pi$ phase boundary corresponds to the nucleation of a semifluxon, this "vortex phase" is, in fact a collection of semifluxons formed at the $0 - \pi$ barrier boundaries. Another possible reason for the co-existence of the π and 0 phases could be the barrier thickness modulation or/and formation of the Fe clusters. However, taking into account a very homogeneous and flat boundaries (Fig. 1), a pure paramagnetic

---

* Electronic address: ondrej.vavra@savba.sk; Present address: Institut für experimentelle und angewandte Physik, Universität Regensburg, D-93025 Regensburg, Germany

[1] B.D. Josephson, Phys. Lett. **1**, 251 (1962).
[2] T. Kontos, M.Aprili, J.Lesueur, F. Genet, B. Stephanidis, and R. Boursier, Phys. Rev. Lett. **89**, 137007 (2002).

[3] V. V. Ryazanov, V. A. Oboznov, A. Y. Rusanov, A. V. Veretennikov, A. A. Golubov, and J. Aarts, Phys. Rev. Lett. **86**, 2427 (2001).

[4] V. V. Ryazanov, V. A. Oboznov, A. S. Prokofiev, V. V. Bolginov, and A. K. Feofanov, Journ. Low Temp. Phys. **136**, 385 (2004).

[5] L.N. Bulaevskii, V.V. Kuzii, and A.A. Sobyanin, JETP Lett. **25**, 290 (1977).

[6] A.I. Buzdin, L.N. Bulaevskii, and S.V. Panyukov, JETP Lett. **35**, 178 (1982).

[7] Y.S. Barash and I.V. Bobkova, Phys. Rev. B **65**, 144502 (2002).

[8] M. Fogelström, Phys. Rev. B **62**, 11812 (2000).

[9] O. Vávra, Š. Gaži, J. Bydžovský, J. Dérer, E. Kováčová, Z. Frait, M. Maryško, and I. Vávra, Jour. Mag. Mag. Mat. **240**, 583 (2002).

[10] P. Lobotka, I. Vávra, Š. Gaži, A. Plecenik, and J.Dérer, J. Low Temp. Phys. **106**, 373 (1997).

[11] T.M. Klapwijk, Physica B **197**, 481-499 (1994).

[12] L.N. Bulaevskii, V.V. Kuzii, and A.A. Sobyanin, Solid State Commun. **25**, 1053 (1978).

[13] I. Vávra, J. Dérer, Š. Gaži, and P. Lobotka, in Proceedings of the 7th Int. Symposium on Weak Superconductivity, p. 289, Smolenice, June 1994 (unpublished)

[14] R.G. Mints and V.G. Kogan, Phys. Rev. B **55**, R8682 (1997).

[15] A. Buzdin, Rev. Mod. Phys. **77**, 935 (2005); I.F. Lyuksyutov, V.L. Pokrovsky, Adv. Phys. **54**, 67 (2005).